

Cummins-ORNL\ Emissions CRADA: **NO_x Control & Measurement Technology for Heavy-Duty Diesel Engines**

**Project ID:
ACS032**

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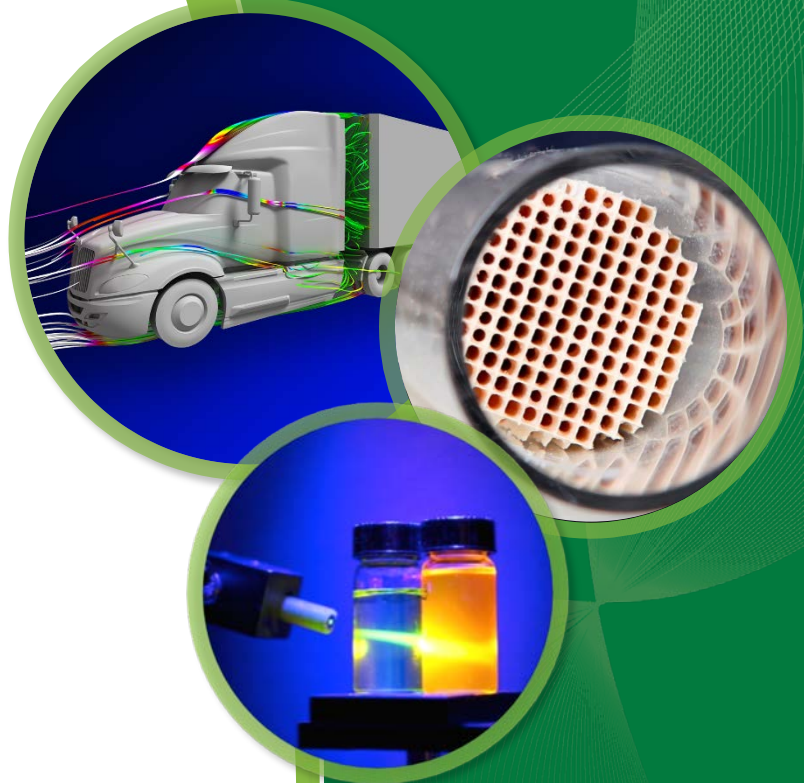
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Oak Ridge National Laboratory
National Transportation Research Center

DOE Vehicle Technologies Office
Annual Merit Review & Peer Evaluation Meeting
June 6, 2017; Washington, DC

DOE Managers:
Gurpreet Singh, Ken Howden, Leo Breton

This presentation does not contain any proprietary, confidential,
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Overview

Timeline

- Year 2 of 3-year CRADA

Budget

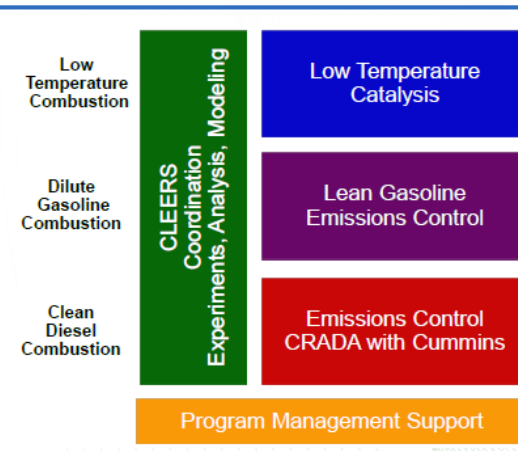
- 1:1 DOE:Cummins cost share
 - In-kind Cummins contribution
- FY17 DOE Funding: \$300k
 - Task3: Cummins CRADA on Diesel Emissions Control
 - Part of ORNL project: “Enabling Fuel Efficient Engines by Controlling Emissions” (2015 VTO AOP Lab Call)

Barriers

- From DOE VT MYPP:
 - 2.3.1.B: Cost-effective emission control
 - 2.3.1.C: Modeling for emission control
 - 2.3.1.E: Emissions-control durability

Partners

- ORNL & Cummins Inc.
- CLEERS

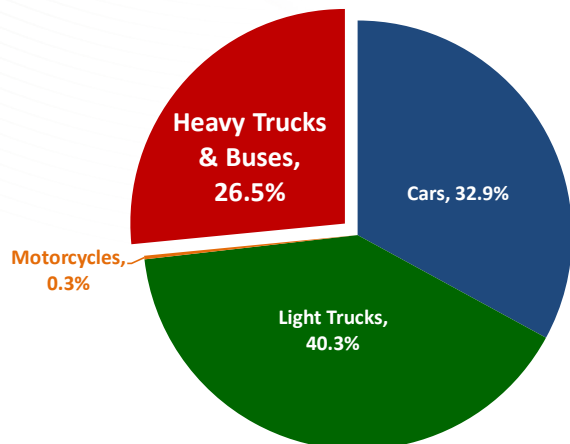


Milestones

FY	Qtr	Milestone & Objectives	Status
2017	1	Compare performance of an SCR catalyst in several states together and with model predictions	complete

Objectives & Relevance

- Fuel economy and emissions reduction remain significant & challenging
- Better catalysts enable engine tuning for greater efficiency vs. emissions
- Efficient, durable & cost effective catalysts enable great benefit



On-Highway Petroleum Use
(Source: Transportation Energy Data Book)

Heavy Duty SCR
Durability Standard:
435,000 miles!

(Source: EPA)

CRADA
Objectives

Catalyst-State
Sensing

Predictive
Models

- Sensing & Control
- Catalyst design models
- OBD: on-board diagnostics
- Engine-efficiency vs. emissions balance

Enabling DOE Goals

- Development cost & speed
- Catalyst cost, control, efficiency & durability
- Fuel economy

CRADA Approach

CRADA Team

- Planning & Goals
 - Catalyst state & control
 - Develop robust predictive models
- Approach
 - Catalyst materials
 - Analytical methods
 - Experiment design

Cummins

- Samples
- Ageing
- Modeling

ORNL

- Analytical Methods
- Measurements
- Analysis

CRADA Team

- Interpretation
 - Basic & Practical
 - Catalyst state
 - Control
 - Modeling
- Next steps
- Reporting

Clean, Fuel-Efficient, Durable
Engines in the Marketplace



Cummins Development

- Catalyst-State Assessment
- Better Control & OBD
- Improved models & design tools, size, cost, durability, performance

**Methods & Insights
Shared with Community**

CRADA Approach

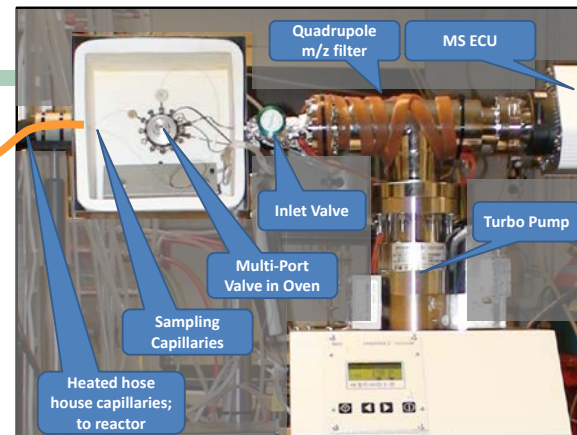
Catalyst Samples



- Commercial Cu/CHA
- Cummins brings supplier knowledge
- Representative field-aged samples
 - Typical of 100s of field-use samples

FTIR

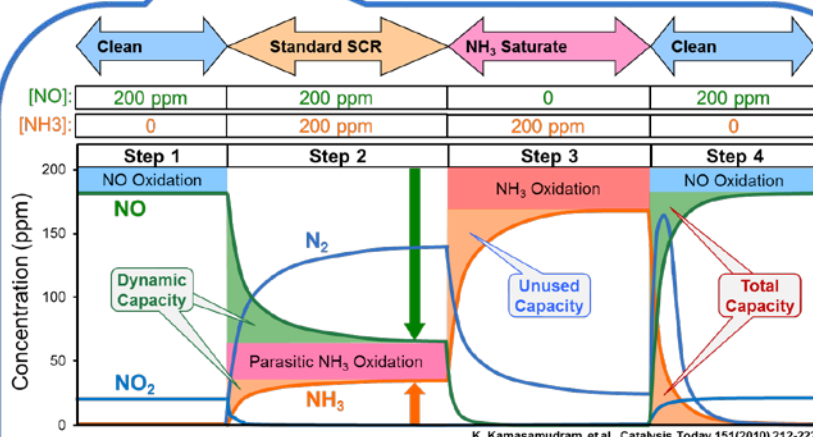
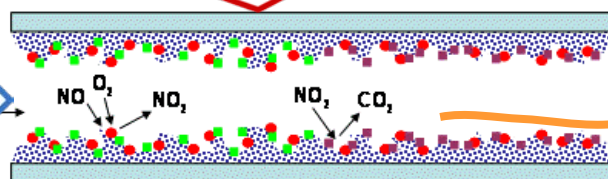
- Effluent species



SpaciMS

- Spatial & temporal mapping
- NO, NH₃, N₂, NO₂, N₂O, other
- 1/16, 1/8, 1/4, 1/2, 3/4, 1L...locations

Gas Mix In



4-Step & Other Protocols

- Different T & species combinations
- Steady state & transient analysis
- Probes specific reactions & functions

Predictive Catalyst Models

- Cummins brings extra-CRADA knowledge
- CRADA focuses on data for critical assessment & insights for improvement

Enabling CRADA Contributions

- Functional relationships & ageing impacts
 - Insights for improved modeling & state sensing
- Catalyst-state sensing
 - Sensing, OBD, efficiency, durability
- Robust & accurate predictive models
 - Design, efficiency, control, durability

Technical Progress: Summary

- **Building on 2016 Progress**

- Assessed applicability of functional insights across range of catalyst states
- Pulse-response method for catalyst-state assessment
- Steady state spatially distributed assessment of Cummins SCR model

- **Transient Assessment of Cummins SCR Model**

- Robust spatiotemporal assessment via 4-Step Protocol SpaciMS data

- **Experimental Characterization of Conversion Inflections**

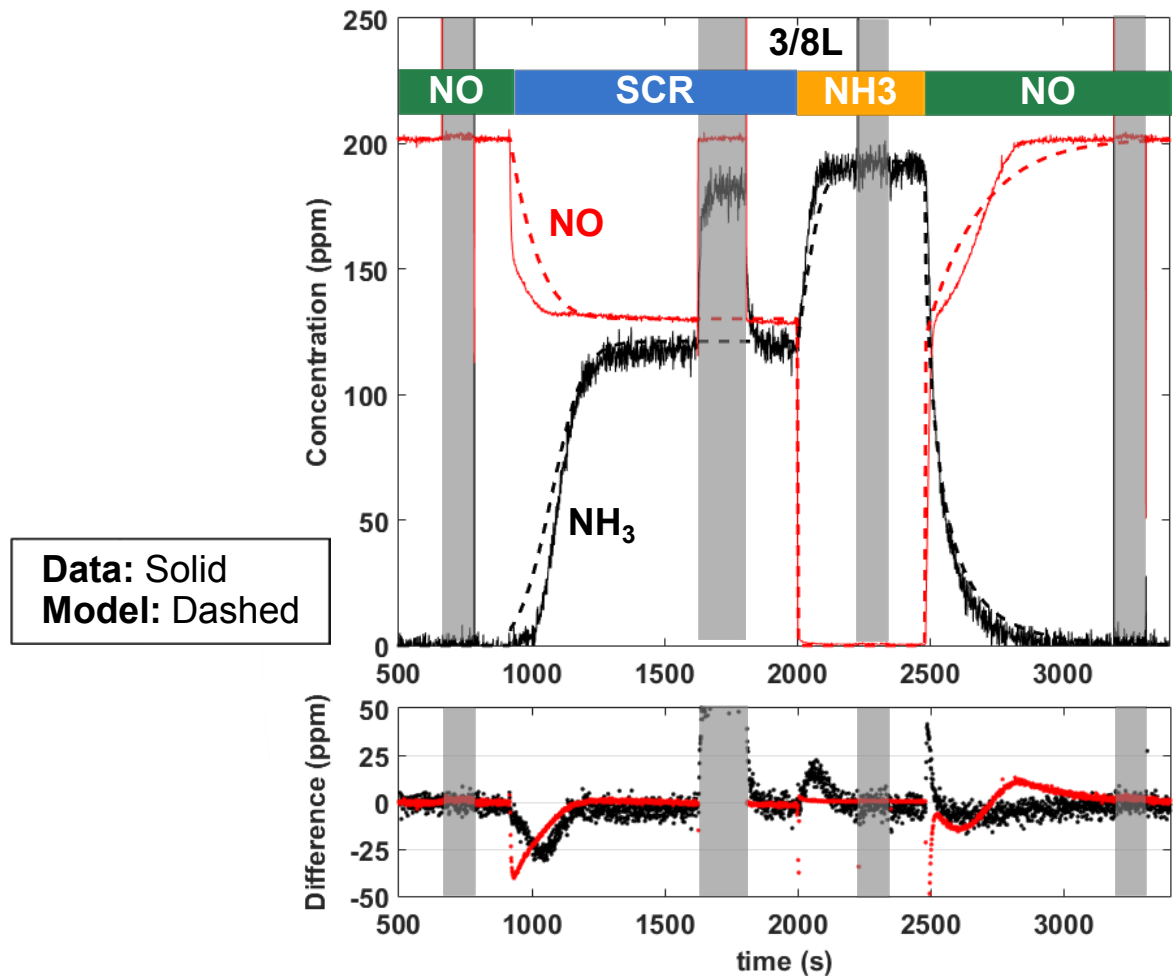
- Variations with catalyst state and reaction conditions
- Common threshold values and features

- **Modeling of Conversion Inflections**

- Insights for improving catalyst model
- Pathway for inflection-based catalyst-state assessment

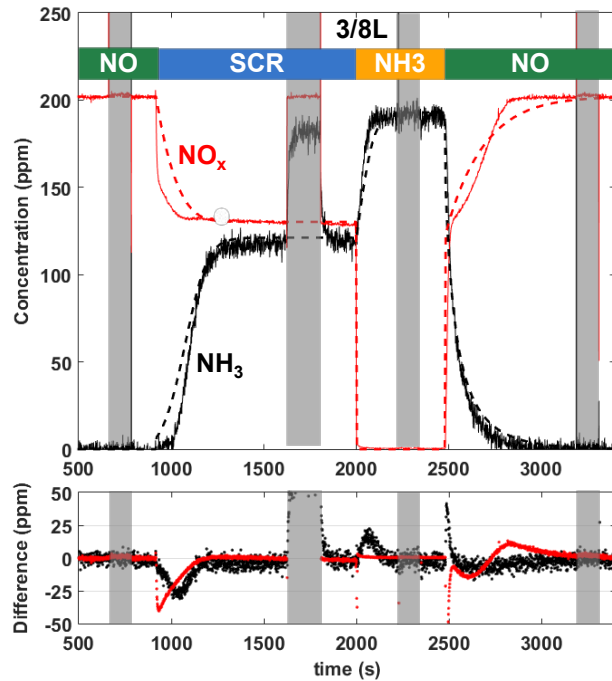
Model Predicts Major Transient Performance Features

- Commercial Cu/CHA; field aged; 300°C; 200ppm Standard SCR; 4-Step protocol

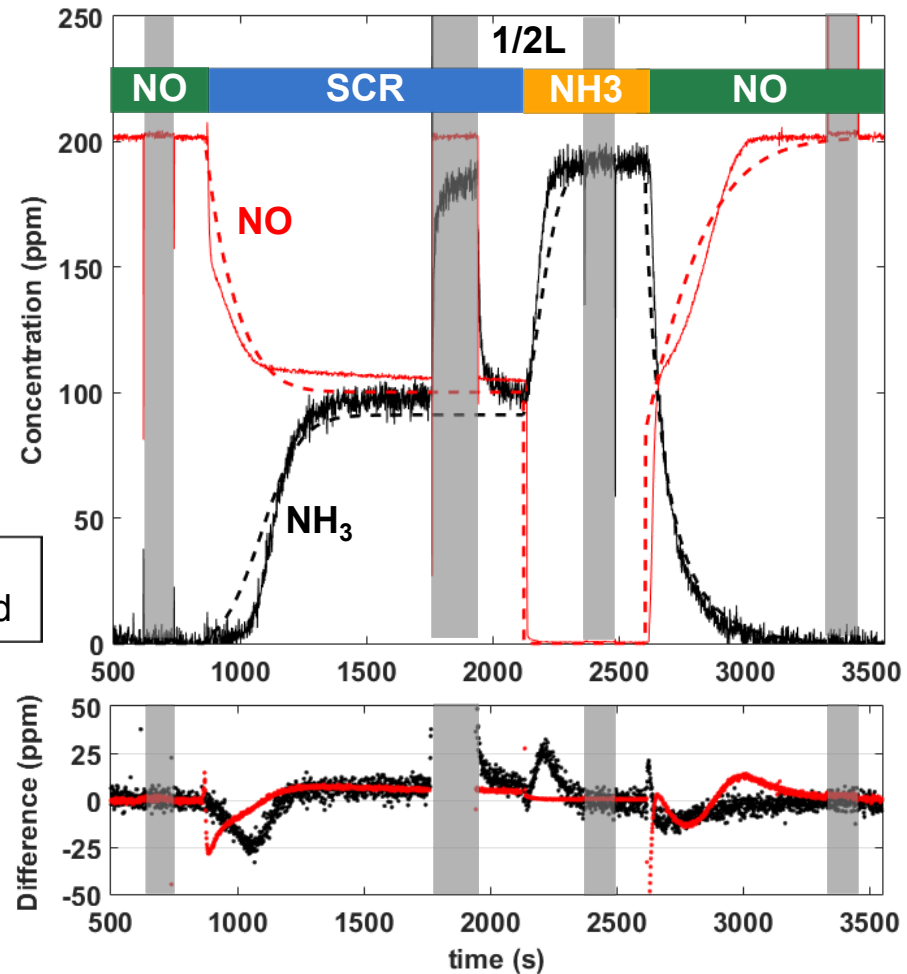


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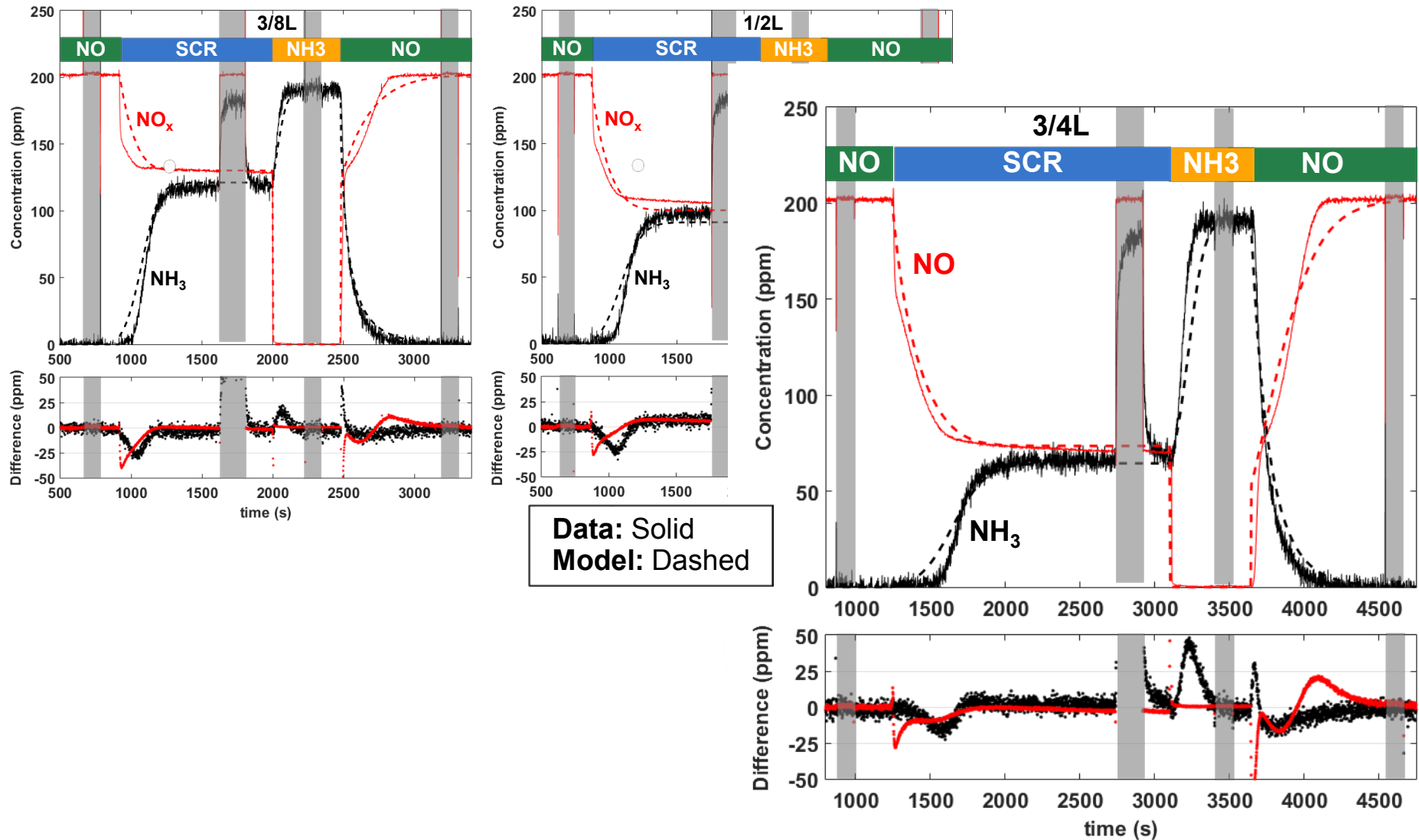


Data: Solid
Model: Dashed



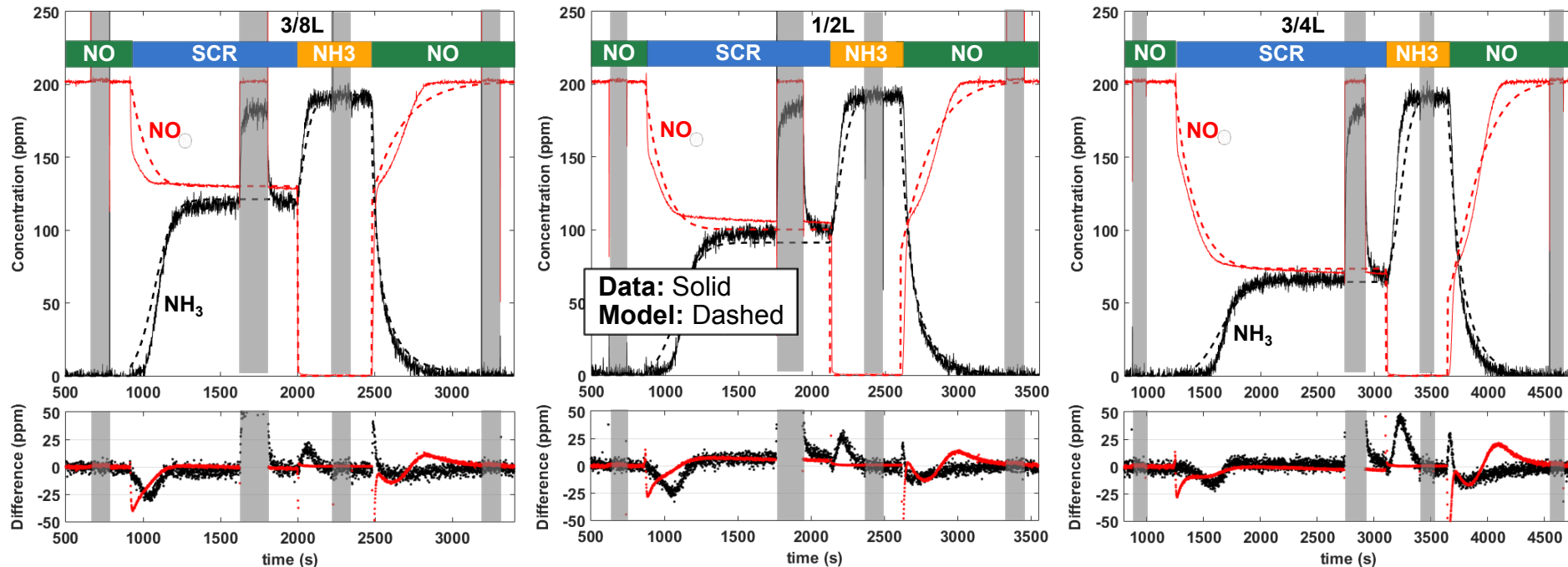
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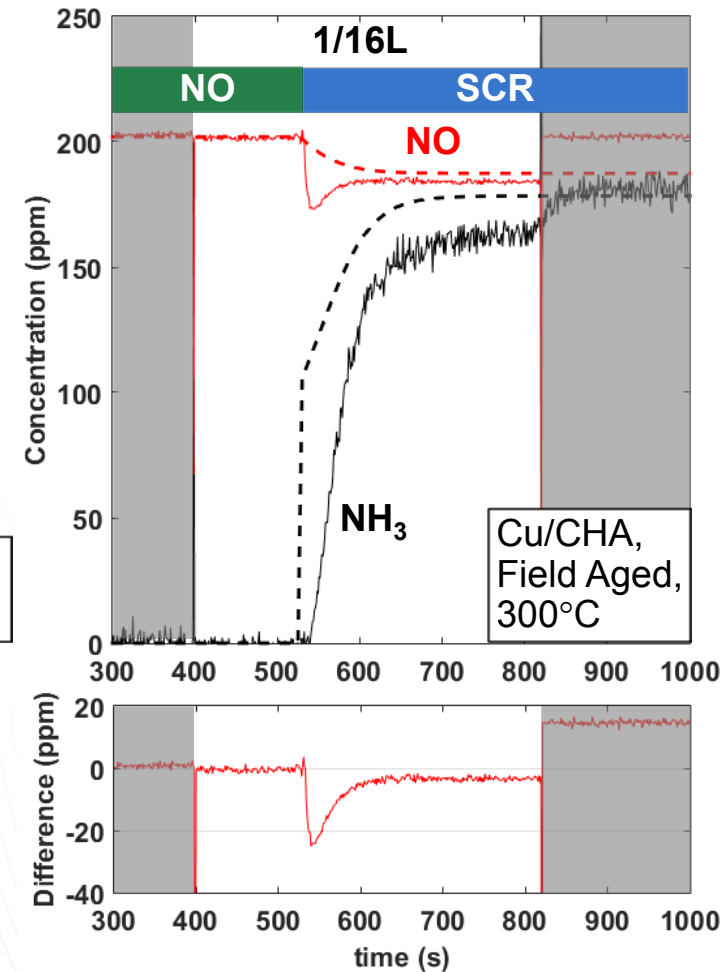
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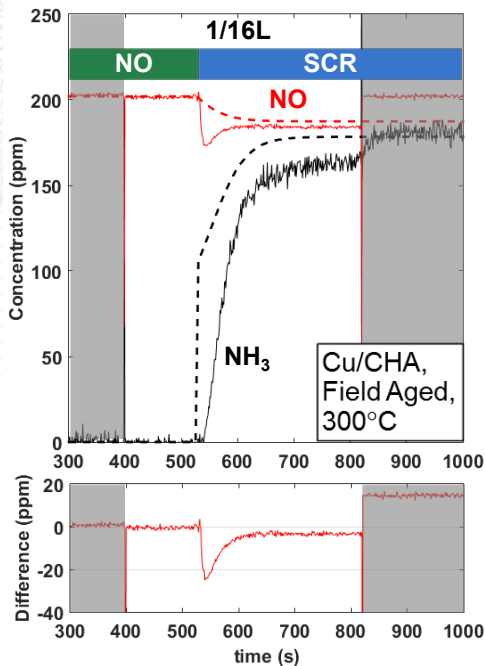
- Model follows major features of each protocol transient in catalyst middle & back
 - Model captures fast initial Step-3 & Step-4 onset transients
 - Slower Step-4 onset transient not as well predicted
- Differences suggest possibility to improve model accuracy & robust nature
 - Initial work focusing on SCR-onset transient
- Spatiotemporal intra-catalyst data enables much more robust & accurate models
 - More extensive calibration & validation data sets

NO Conversion Inflections Not Captured by Model

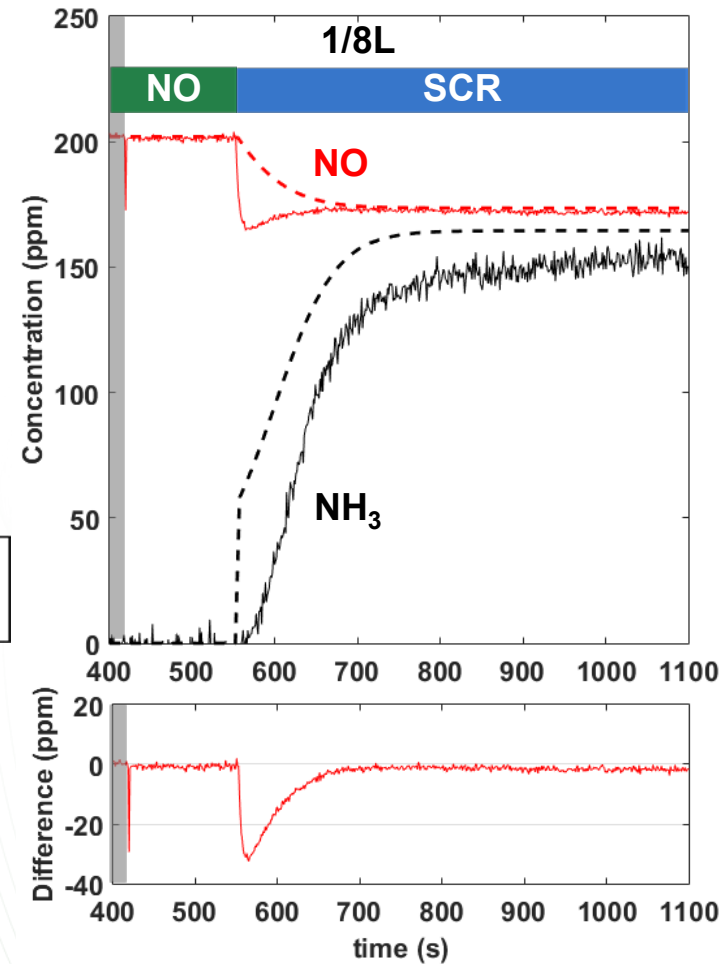


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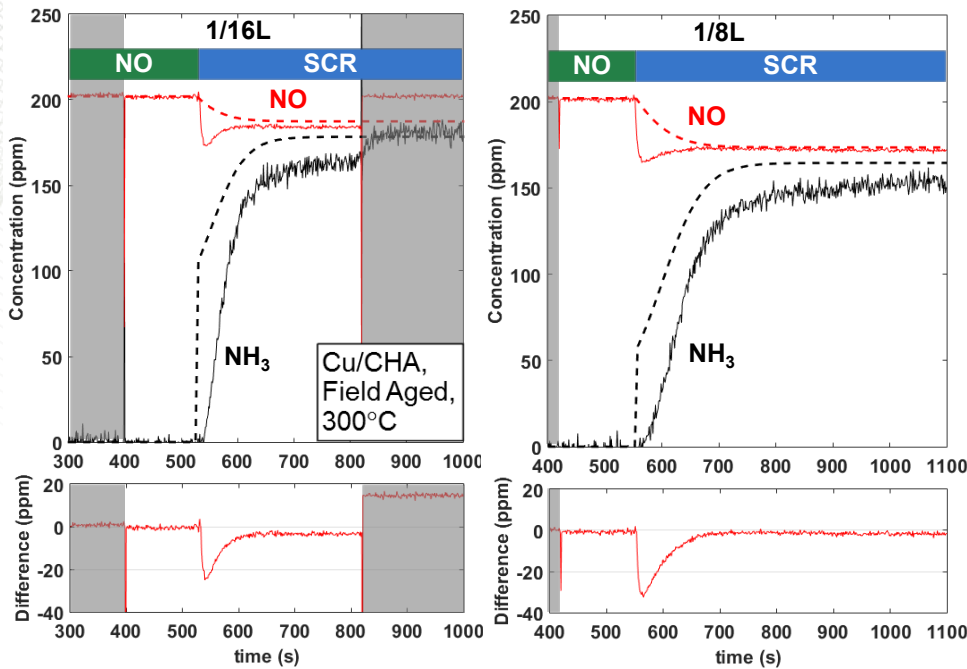
NO Conversion Inflections Not Captured by Model



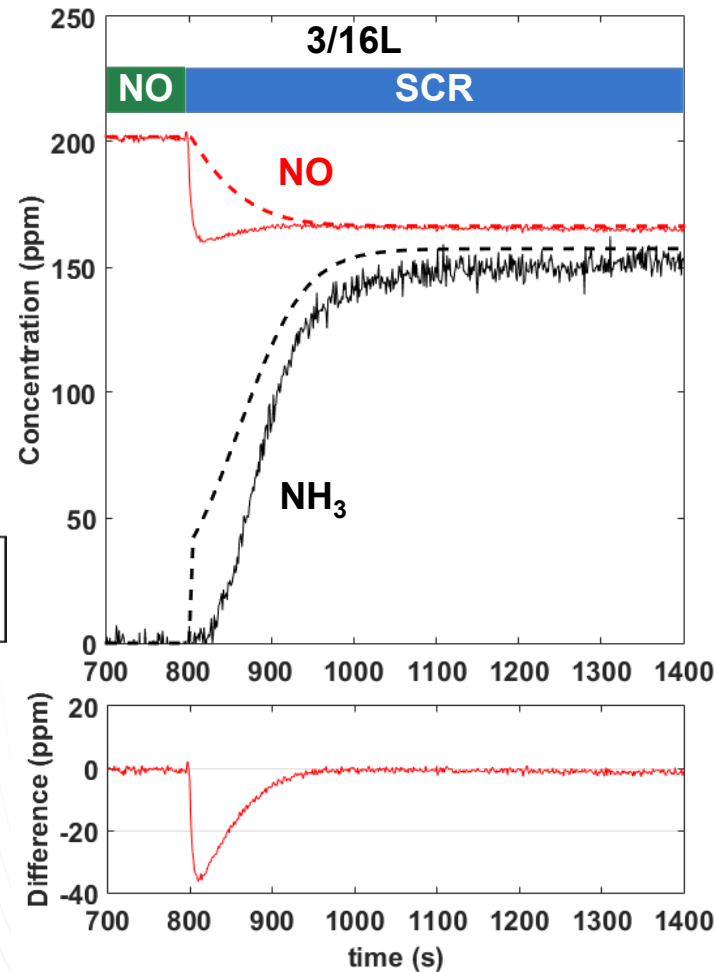
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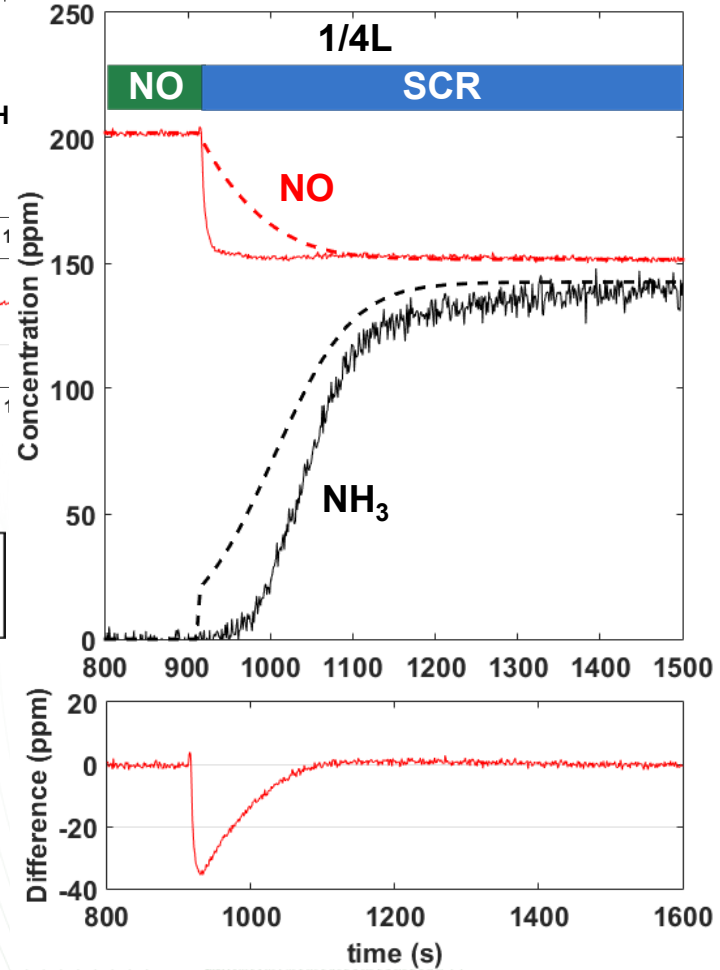
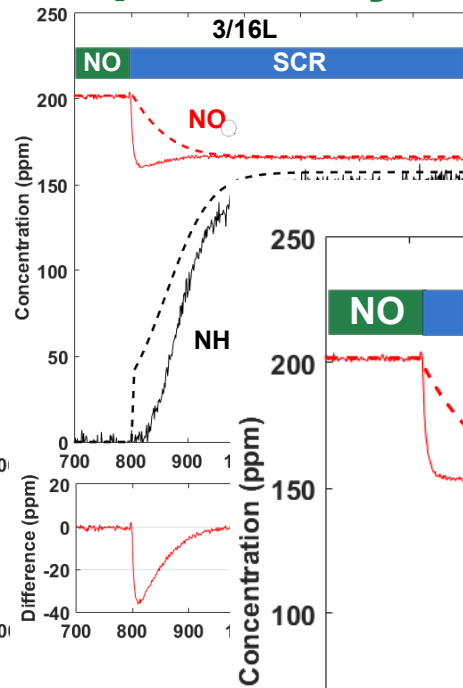
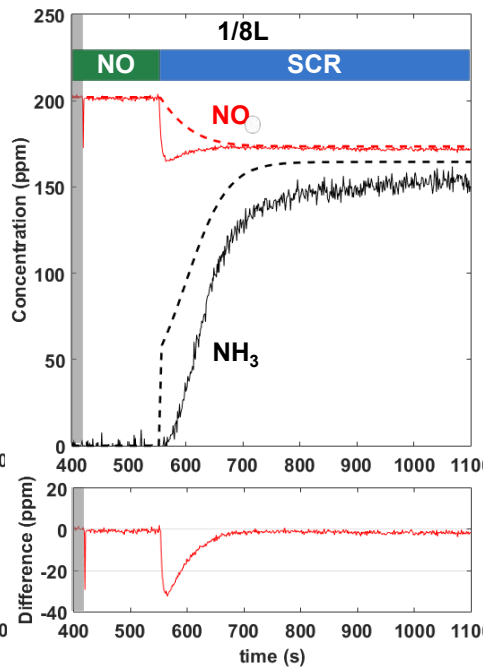
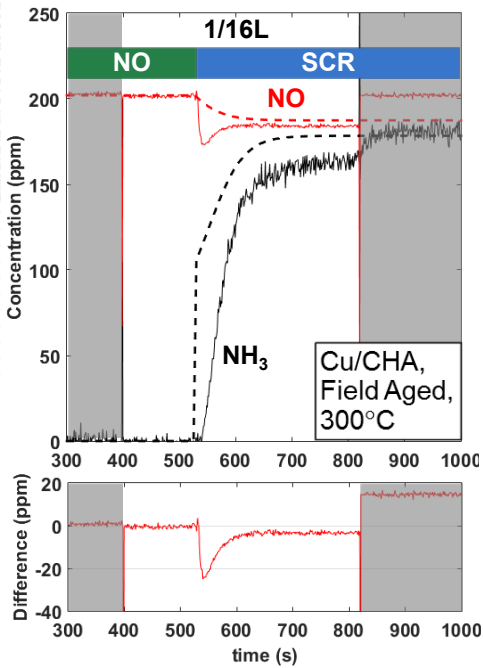
NO Conversion Inflections Not Captured by Model



Data: Solid
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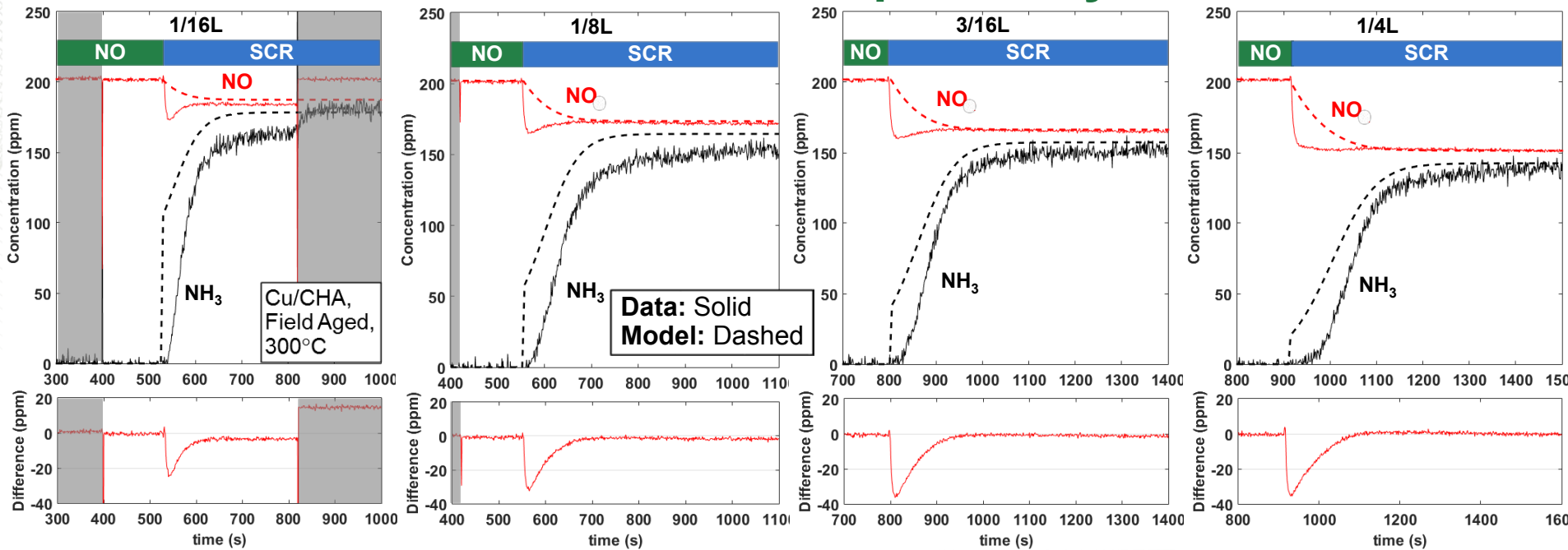


NO Conversion Inflections Not Captured by Model



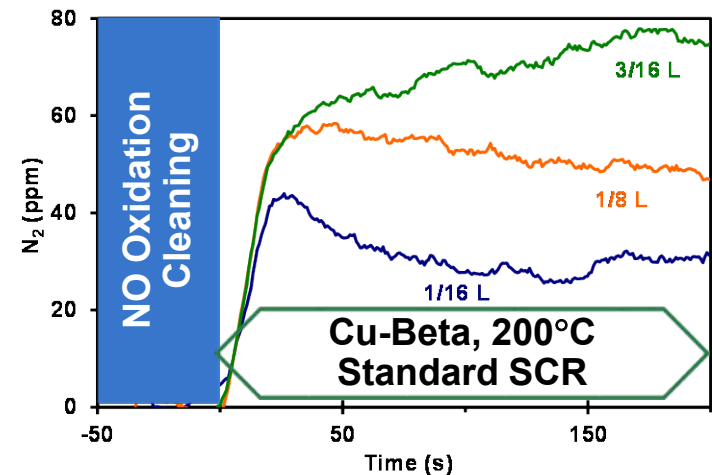
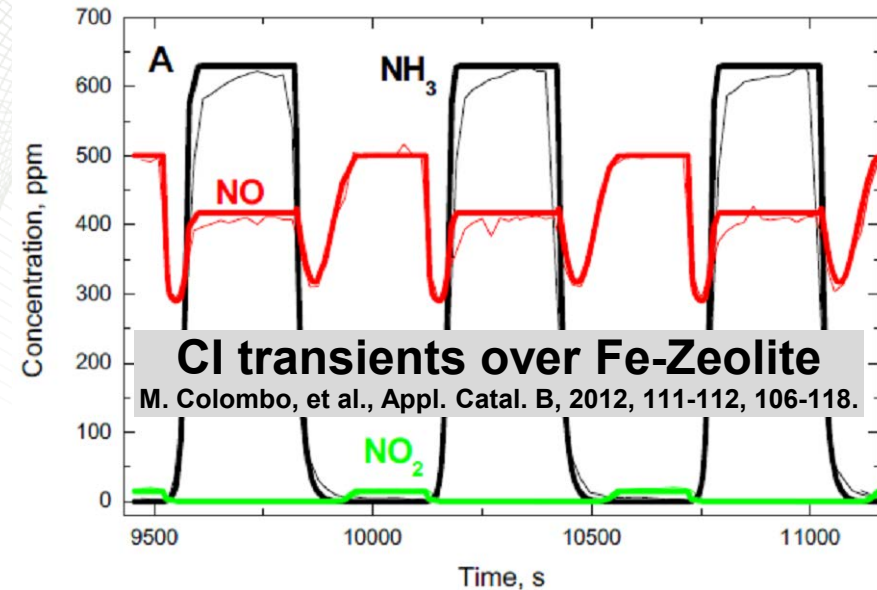
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Model: Dashed

NO Conversion Inflections Not Captured by Model



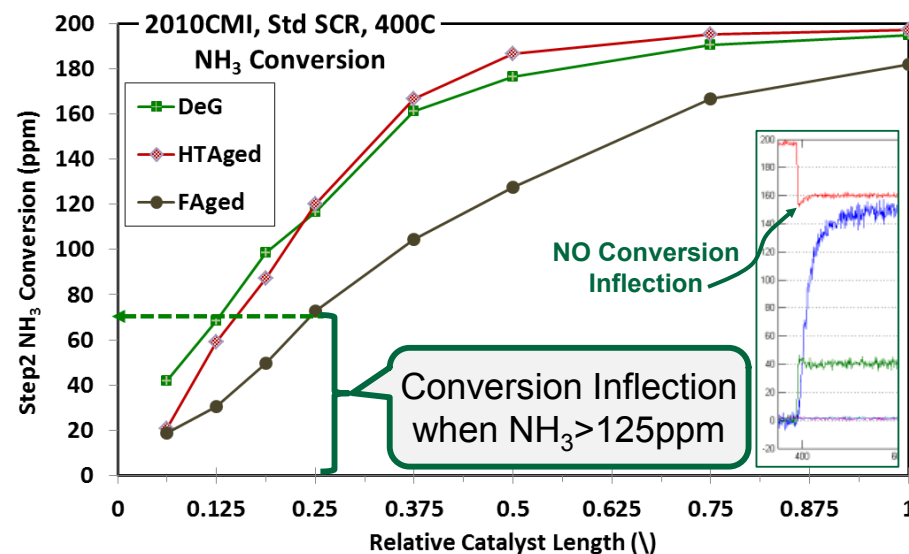
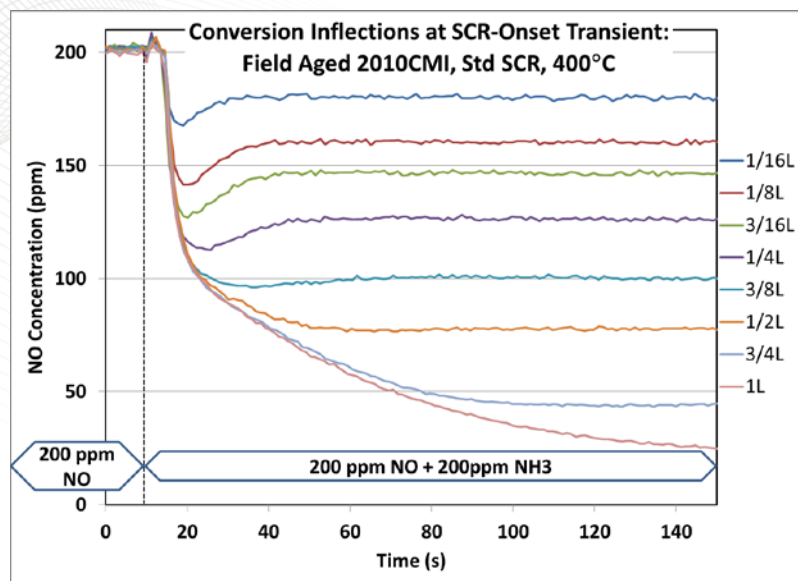
- NO & N₂ inflections occur in catalyst-front regions
 - Greatest at front where NH₃ is high
 - Inflections progressively decay into catalyst as NH₃ (& NO) is consumed
- SpaciMS data allows identification of model limitations not apparent in effluent
 - Provides opportunity to improve catalyst-model performance
- Conversion inflection may provide information regarding catalyst state
 - A naturally occurring alternate to the Pulsed-Response Method

Conversion Inflections Reported for Fe- & Cu-SCR Catalysts



- Conversion Inflections (CI) have been observed over Fe-zeolite SCR catalysts
 - Caused by dynamic NH_3 coverage buildup to an inhibited state
 - Conversion peaks and degrades as coverage grows through optimum value
- We routinely observe CI in front region inside Cu-zeolite SCR catalysts
 - Observed for Cu-Beta ('12 AMR presentation), and in commercial Cu/CHA
 - Most pronounced in front regions where NH_3 concentration is high
 - Not apparent in effluent, thus not broadly known to exist
- Distributed CI transients useful for model critical assessment and improvement

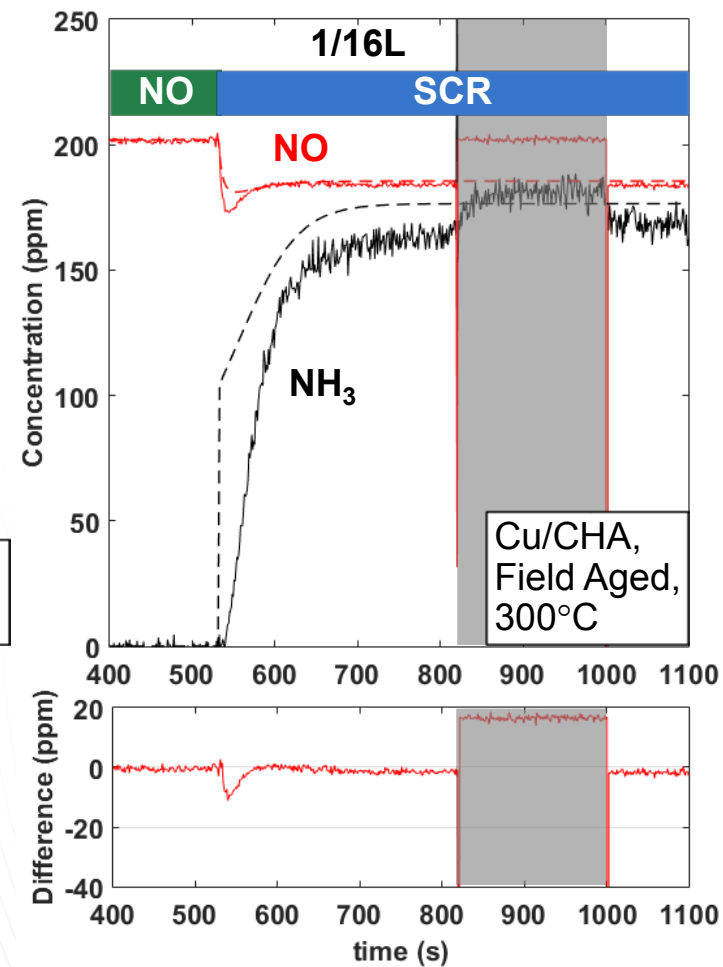
Conversion Inflections may provide Measure of Catalyst State



Partridge, et al., 2015 CLEERS Workshop

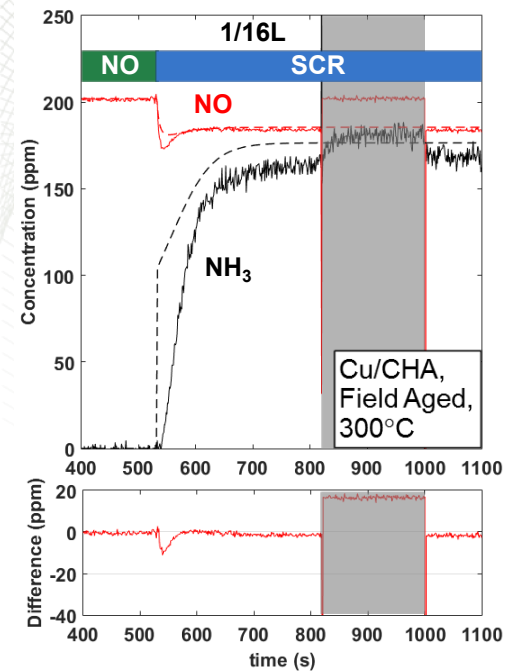
- CI transients appear to vary systematically with conditions and catalyst state
 - Exist deeper into catalyst with lower conversion; due to T, composition, age, etc.
 - Observed above a consistent temperature-dependent NH₃ threshold
- Relevant to catalyst-state assessment, control & OBD
 - E.g., correlate CI timing or location with aged state
 - Feedback for adjusting urea dosing, or model-based control parameters
- Need a better understanding of CI transient nature
 - Study over a range of temperatures, composition & aged states

CI Provides Insights to Improve Catalyst Model

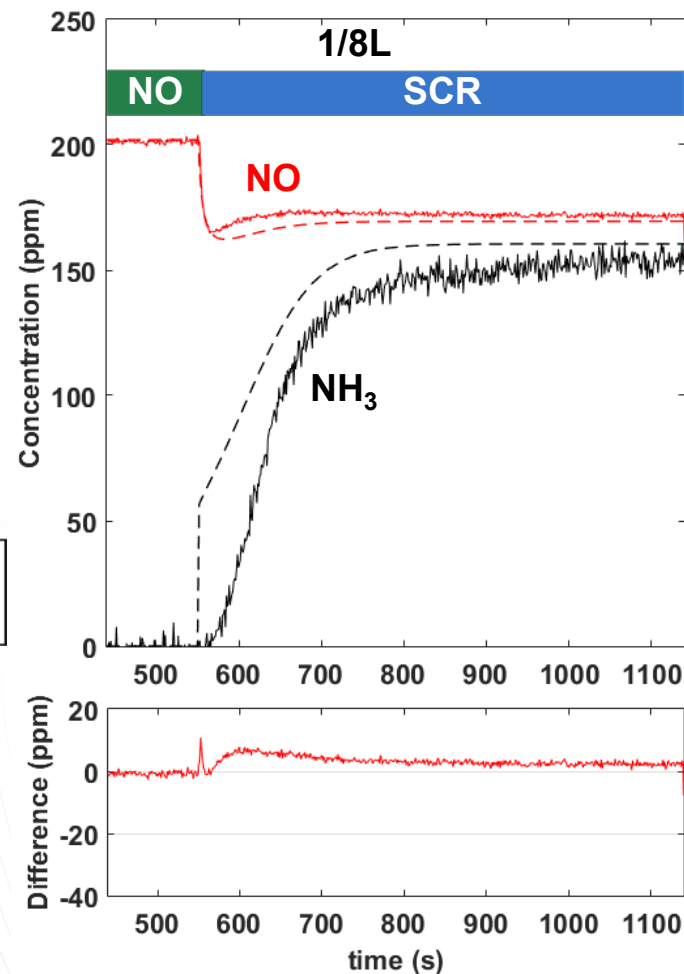


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CI Provides Insights to Improve Catalyst Model



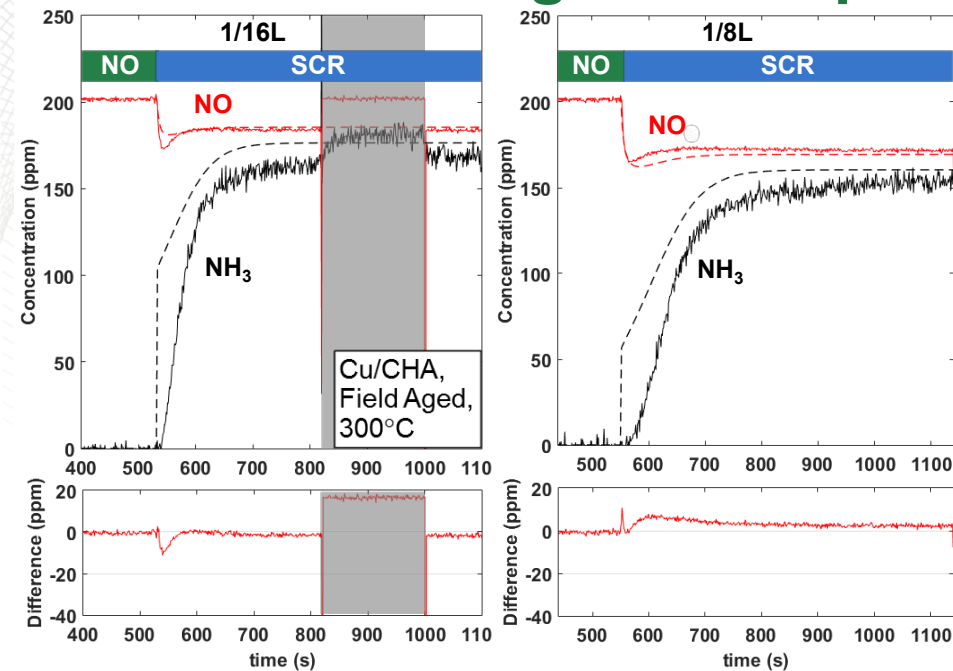
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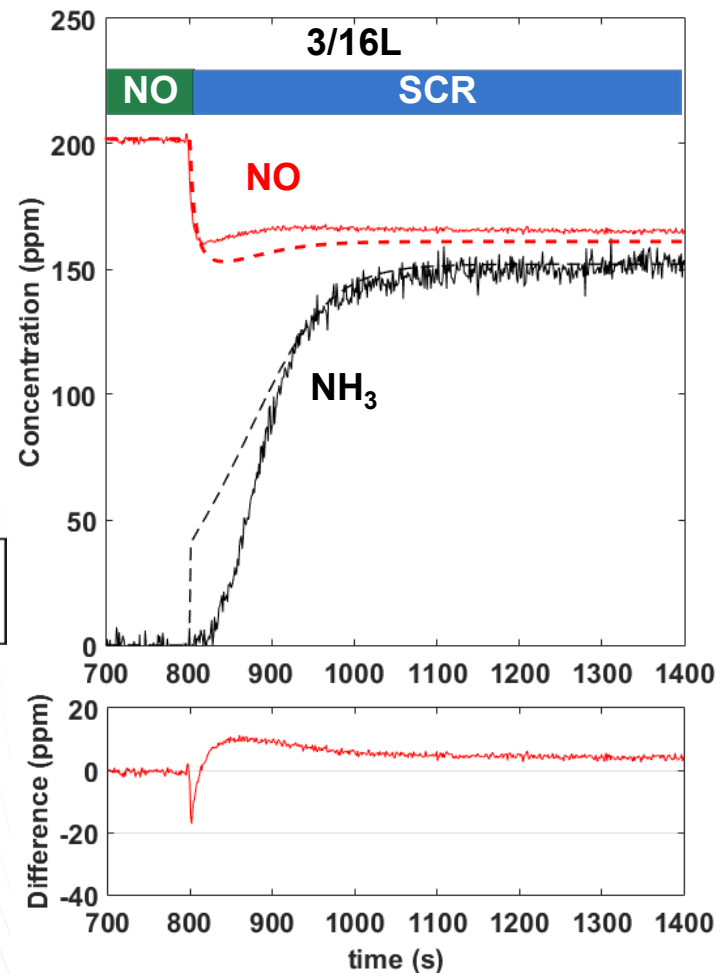
OAK RIDGE
National Laboratory

NATIONAL
TRANSPORTATION
RESEARCH CENTER

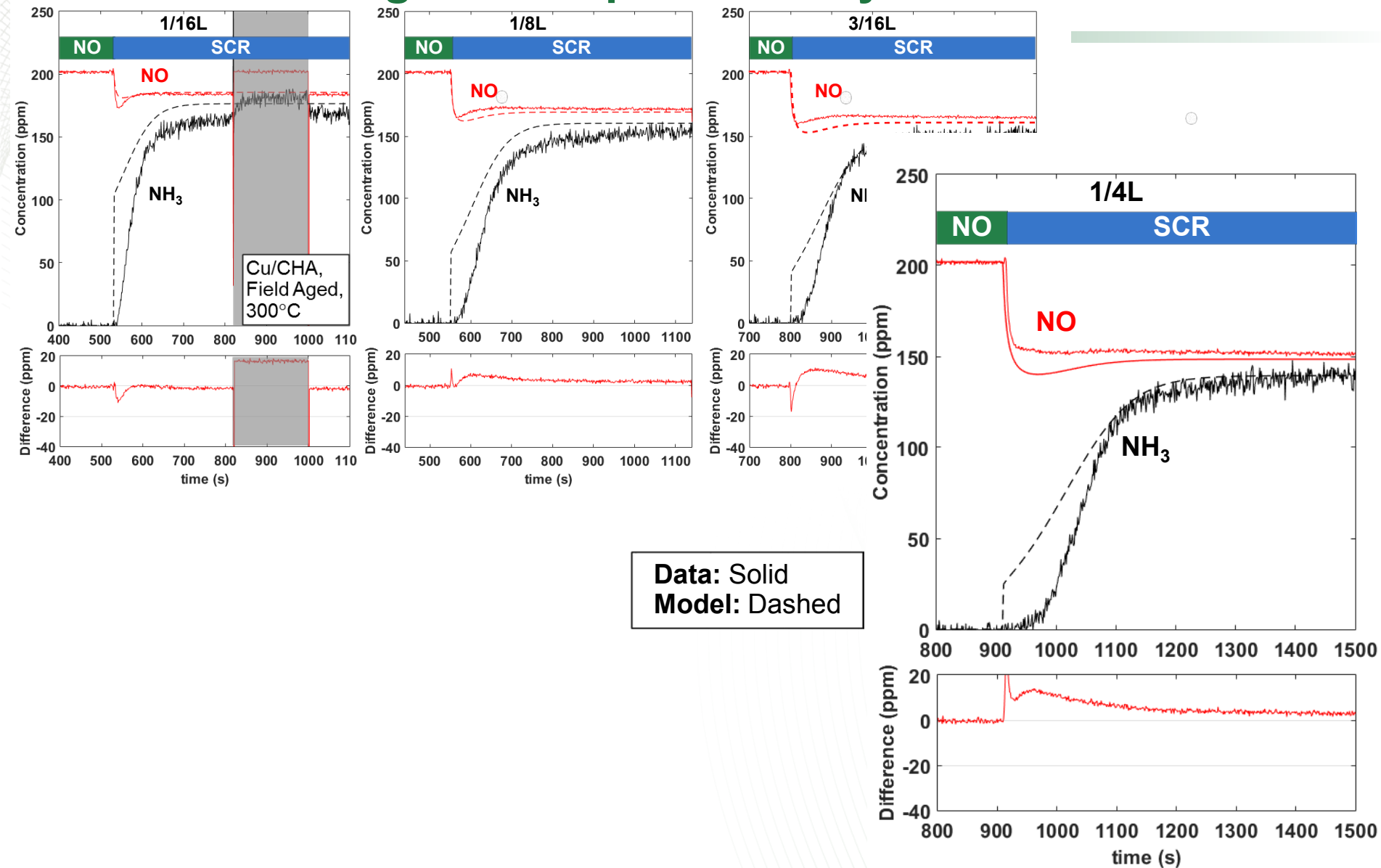
CI Provides Insights to Improve Catalyst Model



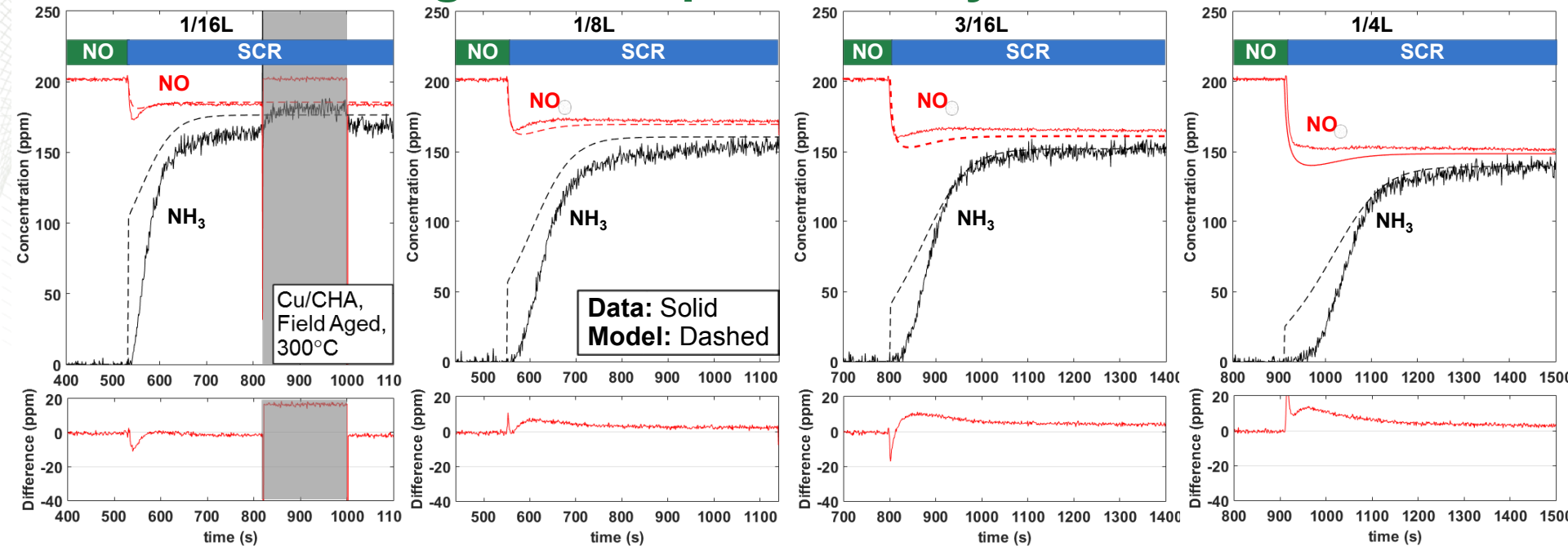
Data: Solid
Model: Dashed



CI Provides Insights to Improve Catalyst Model



CI Provides Insights to Improve Catalyst Model



- Results differ depending on how the inhibition term is formulated
 - Only one inhibition model shown here
 - Including CI in model improves accuracy (*ca. 30-40% lower RMS difference*)
 - Neither model formulation accurately predicts protocol transients
 - Suggests more complex reaction pathway
- Need a better understanding of the CI phenomenon
 - Basic insights for understanding how to improve model
 - Practical insights for catalyst-state assessment and on-road control
 - Study over a range of temperatures, composition & aged states

Remaining Challenges & Future Work

Area	Remaining Challenges	Future Work
Catalyst Knowledge	<ul style="list-style-type: none">▪ Nature of conversion inflections▪ Variation with catalyst state & environmental condition	<ul style="list-style-type: none">▪ Experimental matrix<ul style="list-style-type: none">– Temperature: 210, 300, 400, 500°C, etc.– Gas composition & sequence during transient– Catalyst state▪ Incorporate insights from modeling
Catalyst Modeling	<ul style="list-style-type: none">▪ Robust predictive models<ul style="list-style-type: none">– Analysis-led design– Reduced models for control	<ul style="list-style-type: none">▪ Critical evaluation & tuning based on spatially and temporally resolved performance data▪ Incorporate new knowledge from experiments▪ Adjust internal structure as guided by research
Catalyst Sensing	<ul style="list-style-type: none">▪ Methods for on-road:<ul style="list-style-type: none">– Catalyst-state assessment– Control– OBD: on-board diagnostics	<ul style="list-style-type: none">▪ Mine insights from experiments and modeling for state-assessment pathways▪ Assess pathways via experiments & modeling

Any proposed future work is subject to change based on funding levels

Collaborations & Coordination with Other Institutions

• Cummins

- CRADA Partner, Neal Currier (Co-PI)



CRADA Teamwork & Roles

<u>Cummins</u>	<u>Joint</u>	<u>ORNL</u>
<ul style="list-style-type: none">• Catalyst samples• Field & other ageing• Modeling	<ul style="list-style-type: none">• Planning & Vision• Results interpretation• Monthly+ telecoms	<ul style="list-style-type: none">• Diagnostics• Measurements• Data analysis

• CLEERS (ACS022, Pihl, Wednesday 2:45pm)

- Comparing NH_3 -isotherm data sets and parameters



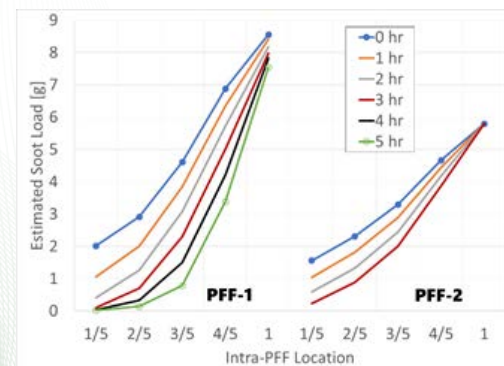
• Politecnico di Milano (Profs. Tronconi & Nova)

- Mechanistic SCR studies (with CLEERS)
- Maria Pia Ruggeri, et al. Topics in Catalysis 59 (2016) 907-912.



• Cummins Partial-Flow Filter Study (Mike Cunningham)

- Spatiotemporal NO_2 utilization & soot loading throughout passive regeneration
- Insights for design and control
- Michael Cunningham, et al. (2017). SAE Technical Paper 2017-01-0988.



Positive Comments:

- “excellent approach,” “targeting key challenges in stepwise manner”
- “unused NH₃ capacity use novel; correlation with pulsed response valuable”
- “collaboration with Cummins excellent and obviously working well”
- “a lot being accomplished for the project budget”

Recommendations:

- Need more extensive field-aged samples and exposure characterization
 - *Focus is not the physics of ageing; expanded work would require additional CRADA resources*
 - *Focus is the practical nature of ageing: functional relations & changes with real-world ageing*
 - *The resulting knowledge has applications for OBD, sensing, control, and modeling*
 - *Knowledge → interpret performance changes → catalyst state & OBD → control → Improve Performance*
 - *Field-aged samples most representative of catalyst in real-world on-highway applications*
 - *Studied samples are suitable to CRADA focus and resources*
- Need to study a modern catalyst
 - *We plan to study a commercial SSZ catalyst*
- Include other characterization methods besides SpaciMS
 - *The team brings capabilities as required to advance tasks and as constrained by resources*
- Limitations of CRADA structure
 - *Cummins incorporates input as appropriate from their industry and academic partners into the CRADA and vice versa; including catalyst suppliers*
 - *The CRADA openly shares diagnostics and analytical methods, functional performance relationships and their variation with ageing, state-sensing insights, capabilities of Cummins’ SCR model*
 - *These are consistent with DOE objectives, and open for use by the broader community*

Category	Score
Approach	3.10
Tech Progress	3.10
Collaboration	3.10
Future Research	3.10
Weighted Average	3.10

Summary

• Relevance

- CRADA work enables improved catalyst knowledge, models, design, OBD & control
- This reduces catalyst system costs & required engine-efficiency tradeoffs
- This in turn enables DOE goals for improved fuel economy

• Approach

- Develop & apply diagnostics to characterize catalyst nature
- Analyze data to understand details of catalyst functions, interrelations & ageing impacts
- Develop catalyst-state-assessment methods & predictive catalyst models

• Technical Accomplishments

- Assessed transient spatially distributed performance of Cummins SCR model
 - Identified opportunity to improve model
- Experimental characterization and modeling of conversion inflections
 - Potential for catalyst-state assessment & enabling more robust predictive model

• Collaborations

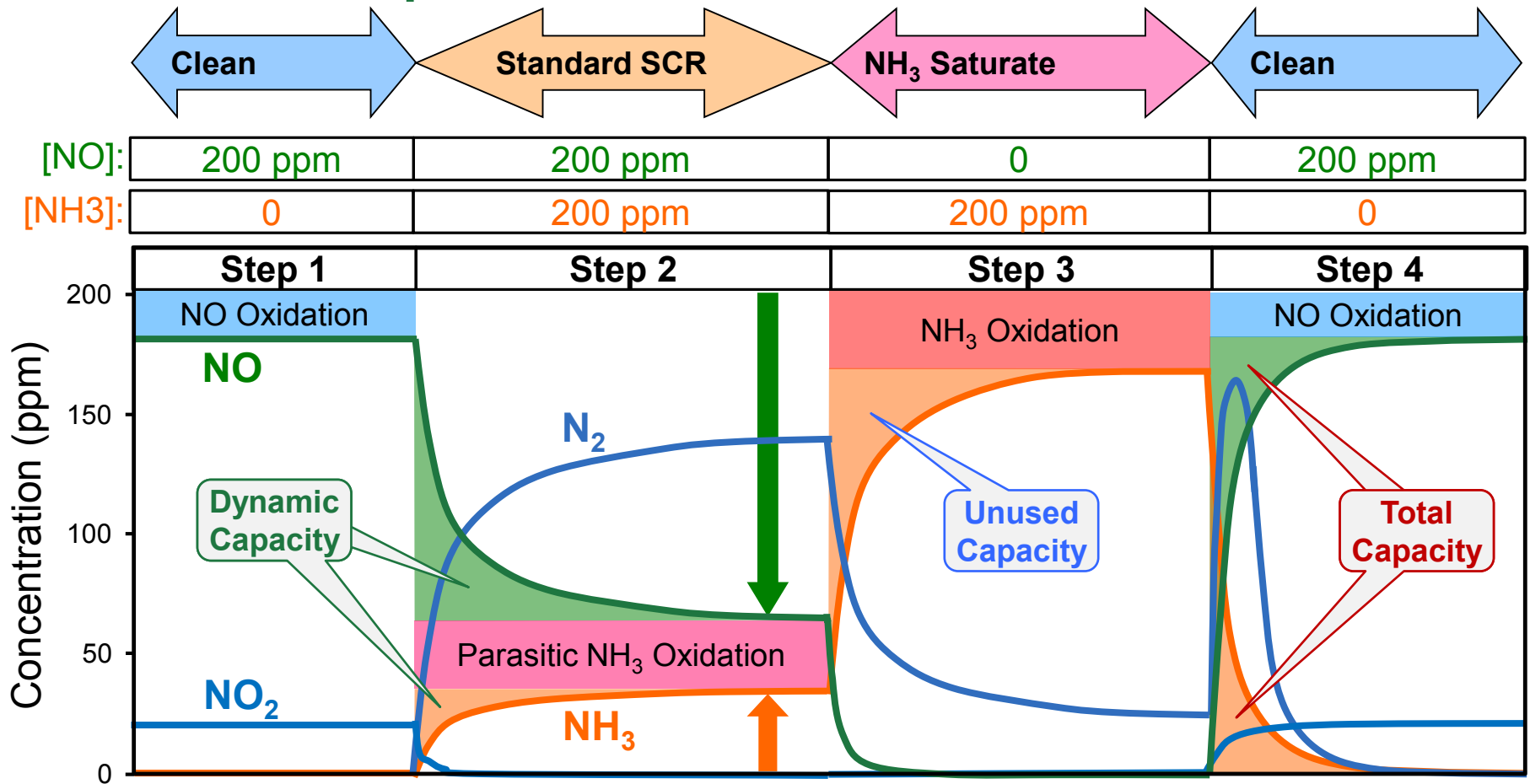
- Multiple collaborations resulting in presentations, publications and advances
- Coordination & collaboration with other DOE projects to maximize benefit

• Future Work (Any proposed future work is subject to change based on funding levels)

- Conversion inflection characterization and modeling at range of conditions
- Mine insights to advance catalyst state, modeling and OBD objectives

Technical Back-Up Slides

Cummins 4-Step Protocol Resolves Reaction Parameters



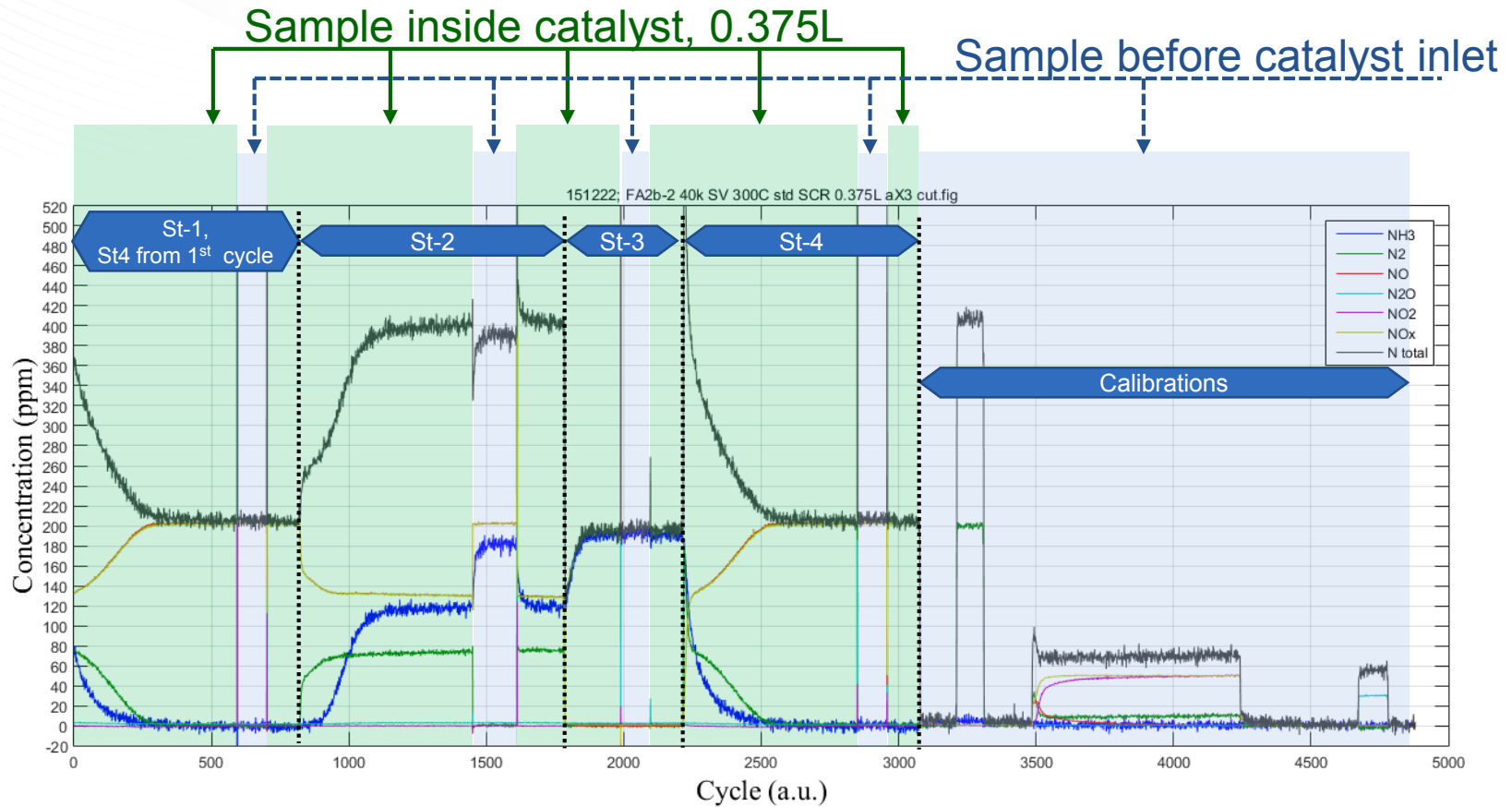
K. Kamasamudram, et al., Catalysis Today 151(2010) 212-222.

- Step1: NO oxidation
- Step2: SS NO_x & NH_3 conversions, Parasitic NH_3 oxidation, Dynamic NH_3 capacity
- Step3: NO_x -free NH_3 oxidation, Unused NH_3 capacity
- Step4: NO oxidation, Total NH_3 capacity

$$\text{Total} = \text{Dynamic} + \text{Unused}$$

Data from 4-Step Protocol

Inlet is sampled in SS region of each protocol step



2016 AMR Technical Progress Slides



Technical Progress: Overview

Enabling SmartCatalyst Systems

Tools & Models for better Design, OBD & Adaptive Control

Impacts of Field-Ageing & Adaption Methods

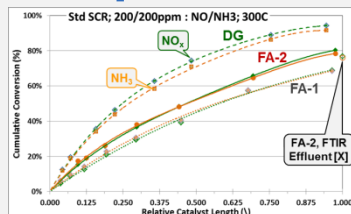
- Field-Aged sample compared to DeGreened
 - Lower SCR conversion and Total NH_3 Capacity
 - Same Dynamic NH_3 Capacity and adsorption energetics
 - Can use same adsorption model with scaling factor

Are insights from FA-1 generally applicable?

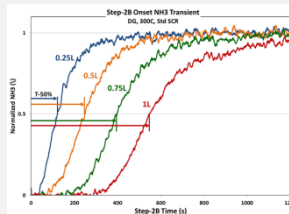
Concept for catalyst-state assessment

Spaci data allows critical model assessment

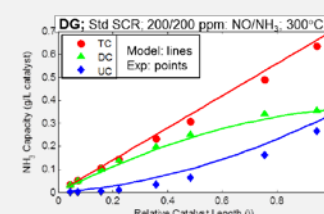
2nd Field-Aged Sample



Method for State Assessment



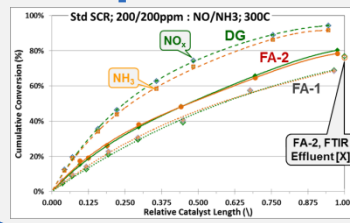
Validate Cummins Predictive Model



FY 2015

FY 2016

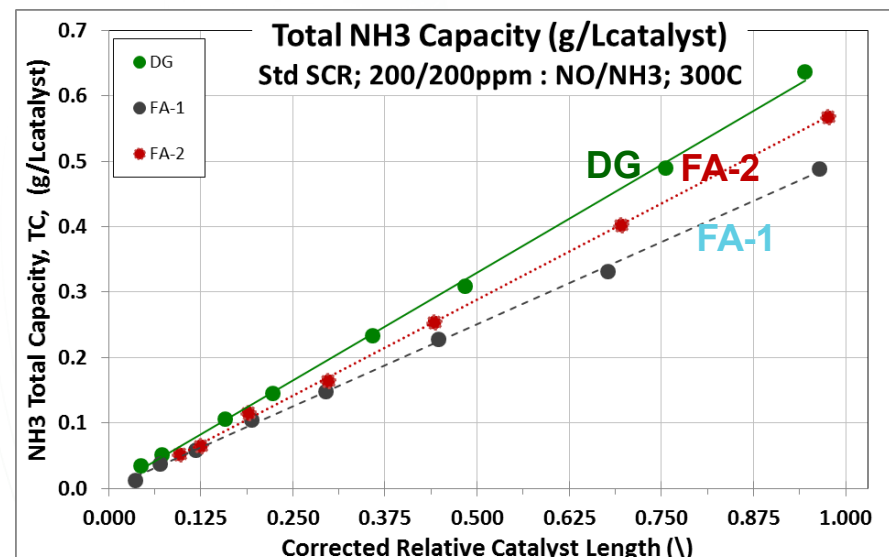
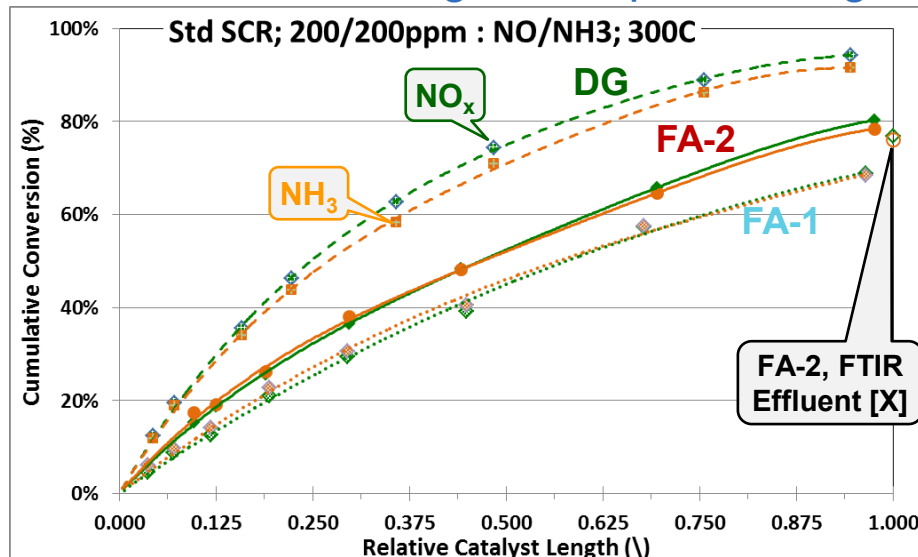
2nd Field-Aged Sample



Tech.Prog.: Similar Impact from two Field-Aged Catalyst Samples

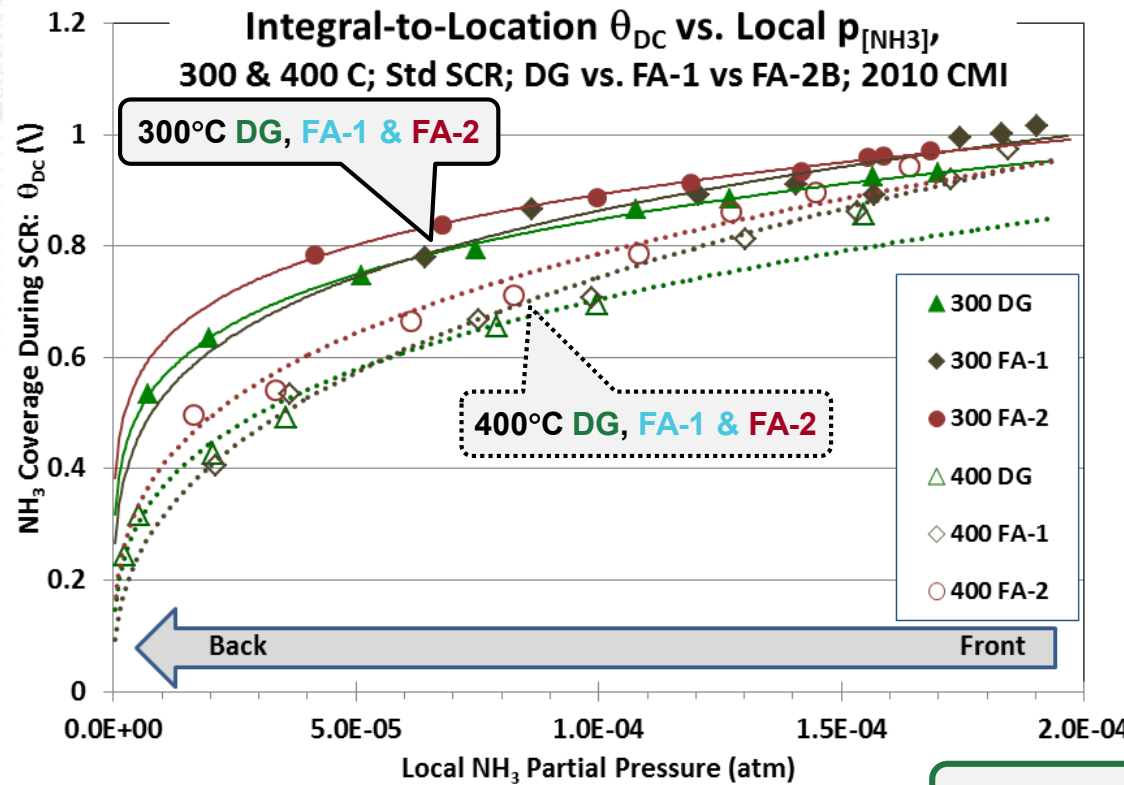
Are FA-1 insights (2015 AMR) applicable to other Field-Aged (FA) samples?

- Will a single predictive model work for different Field-Aged samples?
- Commercial 2010 Cummins, Cu/SAPO-34 SCR catalysts
 - Different field exposures; unknown exposure details; normal ageing profiles
 - Value – real-world on-road use; data to critically assess predictive model
- **Two different field-aged samples show similar performance response**
 - Field ageing degrades conversion & Total NH₃ Capacity (TC)
 - Same capacity utilization correlations (cf. Backup Slides)
 - Does not change absorption energetics (cf. Operando Isotherms)



Similarities suggests FA-1 insights are applicable to other FA samples

Tech.Prog.: Field Ageing does not Practically Impact NH₃ Adsorption Energetics



- **Operando isotherms**
 - Measured under SCR-reaction conditions: $NO_x + NH_3$
 - vs. ‘neat’ studies: without NO_x
- Shape of 2-site Langmuir
 - cf. neat CLEERS studies
- Selective adsorption-site ageing would change shape
 - Δ Energetics \rightarrow Δ Shape

- Similar NH₃ adsorption energetics for three catalyst states: DG, FA-1 & FA-2
 - Same shape at a given temperature
 - Compliments CLEERS work (ACE032)
 - Neat isotherm studies; SSZ & SAPO
 - CRADA provides operando & ageing

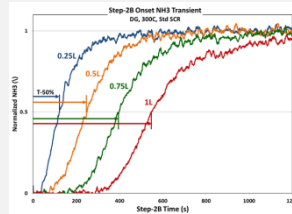
Simplifies modeling of aged samples

- Field ageing reduces number of sites
- But adsorption occurs in same way
- Use same model with scaling factor

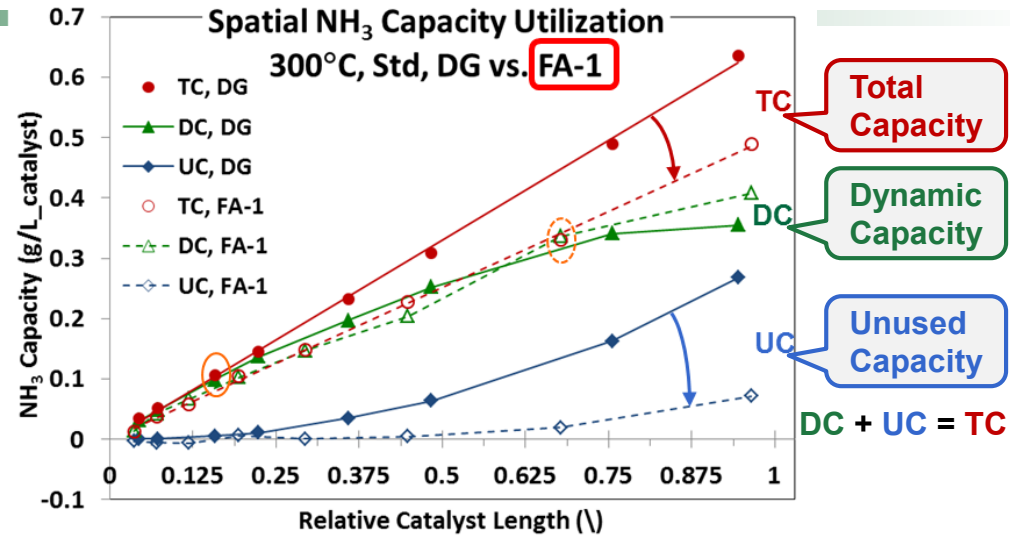
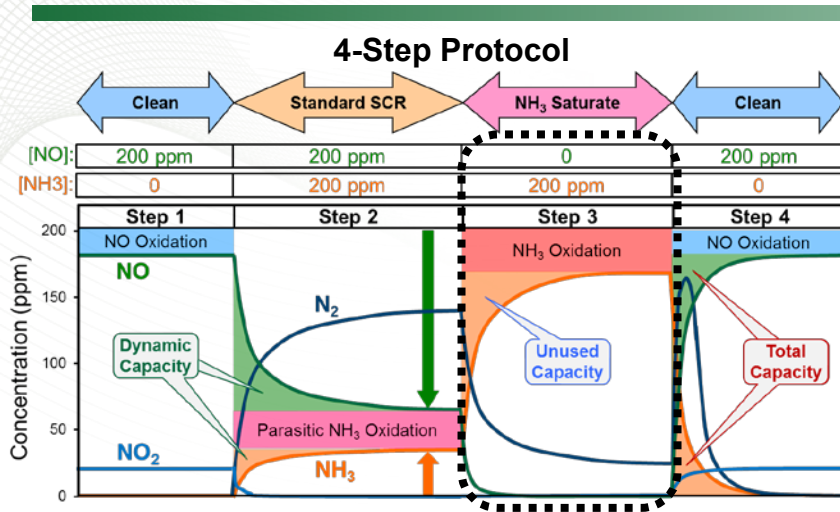
Need to determine catalyst state

- i.e., age-dependent **TC** & scaling factor

Method for State Assessment

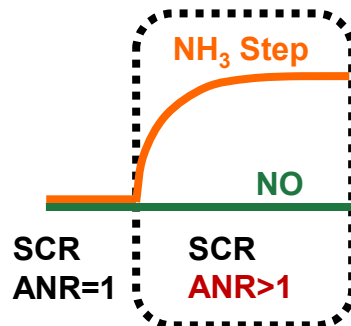


Tech.Prog.: Transient Response Measurement of Catalyst State



- **DC**, **UC** & **TC** capacity components determined from 4-Step Protocol

- **UC** varies with catalyst state
 - **UC** & **TC** variations are related
 - Use to measure catalyst state

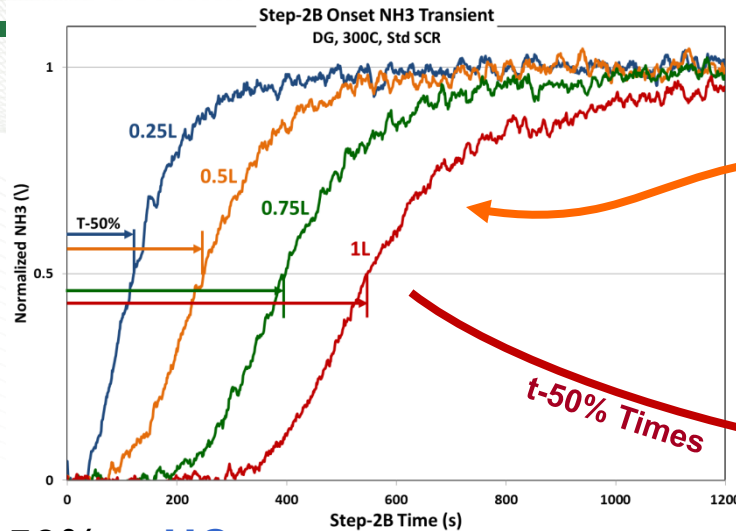


- Protocol not practical for on-road use
- NH₃ step response to probe **UC** variations
 - SCR continues with NH₃>NO
 - Could implement via dosing control

Use for active catalyst-state determination

- Determine factors for feedback
 - e.g., **TC** scaling factor for predictive-model
- Enable OBD & adaptive control

Tech.Prog.: Stepped SCR Dosing to Determine TC Scaling Factor



NH₃ Response

NH₃ Step

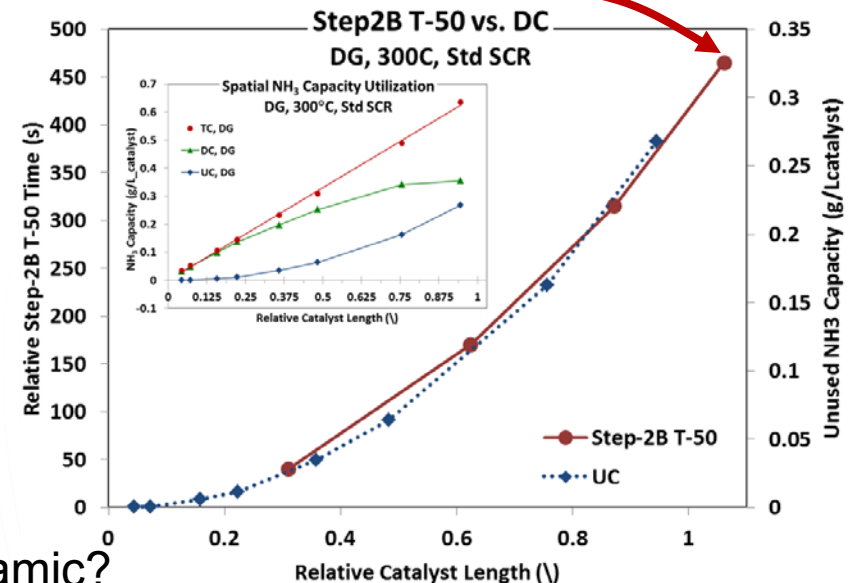
NO

SCR
ANR=1

SCR
ANR>1

- $t_{-50\%} \propto UC$

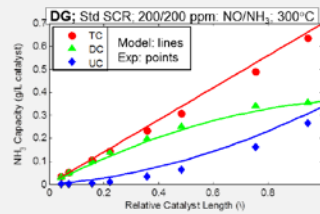
- Varies with ageing; DG vs. FA
- Possible to use other transient features
- Alternate probes might be possible
- Many challenges remaining
 - What pulse & characteristic to use
 - Correlations with catalyst state & factors
 - Use probe pulse or natural drive-cycle dynamic?



Approach for practical catalyst-state assessment

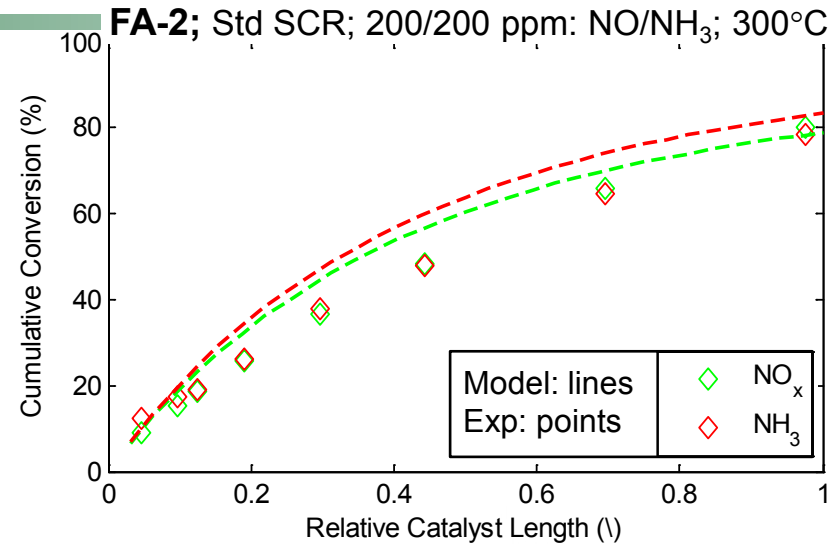
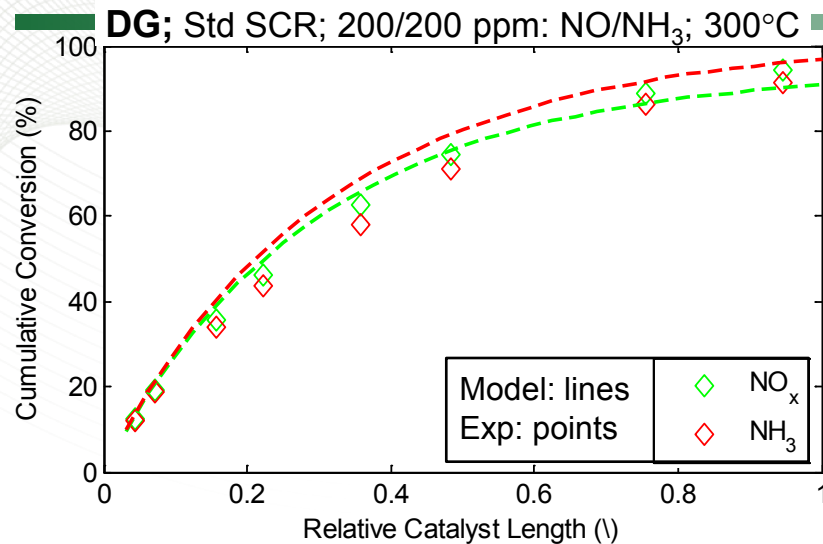
Pathway for thru-life OBD & feedback-enabled adaptive control

Validate Cummins Predictive Model



OAK RIDGE
National Laboratory

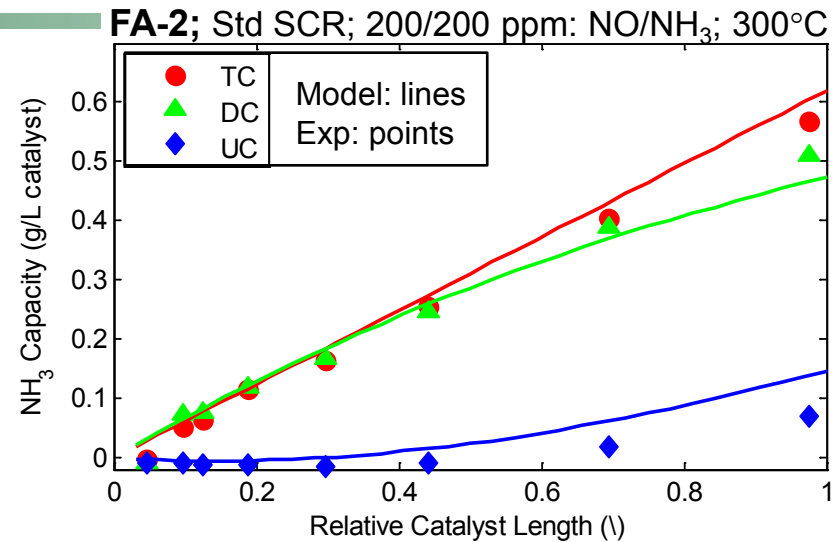
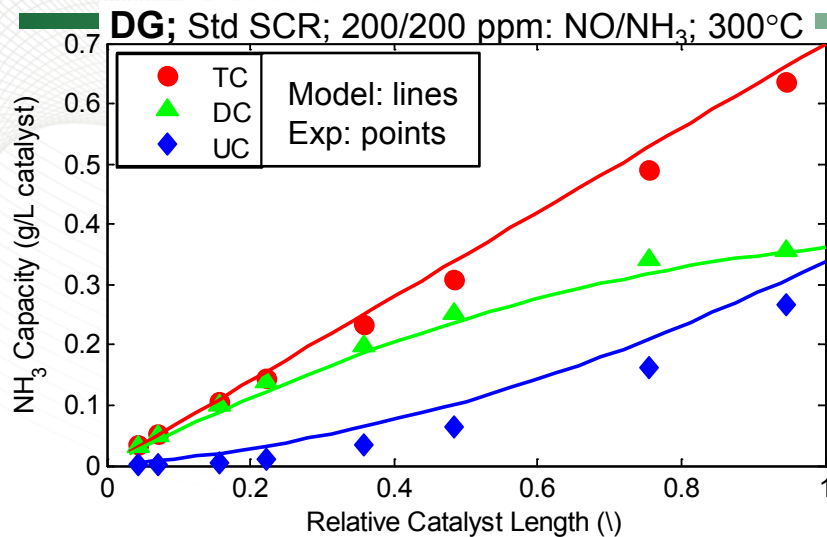
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Goal: develop a predictive model to determine the catalyst's internal state

- Cummins' base predictive model is very complex
 - Many ways to structure & numerous model parameters to fit/determine
 - Developed based on integral DeGreened catalyst measurements
 - Run in AVL Boost
 - Predictive model modifications for FA-2 sample
 - Reduced SCR reaction pre-exponential (*impacts conversion and DC distributions*)
 - Reduced integral TC (*impacts TC & UC distributions*)
 - Model accurately predicts conversion distributions within the catalyst
 - Ability to predict intra-catalyst distributions a rigorous assessment test
- Particularly for catalysts with internal storage functions like SCR and LNT

Tech.Prog.: Validating Cummins Kinetic-Model Structure; P2/2



- Model accurately predicts internal NH₃ capacity utilization
 - In different catalyst states: DG & FA-2
 - Relationships between **TC**, **DC** & **UC** components
 - Supports pathway for catalyst-state assessment; i.e., via **UC** measure

Predictive model rigorously validated vis-à-vis conversion & capacity distributions

- *Confirms that model's internal workings are structured & fit properly*
- *Enhances confidence in its use to design OBD & reduce design margins*

Next step is assessing transient performance